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GASIFICATION OF A CONDENSED MATERIAL IN A HOT GAS

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#### Abstract

A theoretical analysis is presented for the gasification process of a condensed material suddenly exposed to a hot stagnant gas.

When the gasification rate is given by an Arrhenius law the surface temperature rises to the non-gasification jump value upon contact with the hot gases. The convective effect due to surface regression and gasification flow, plus the energy absorbed by the gasification causes the surface temperature to decrease with time. An analytical description of the surface temperature and concentration history is presented, together with closed form expressions for the temperature and concentration profiles for small and large times.

A closed form analytical description is also given for the gasification process under surface equilibrium "Clasius Clapeyron" conditions. In this case the surface temperature remains at a constant jump value, and the temperature and concentration profiles have a similarity form.

## Nomenclature

- P Pre-exponential factor Specific heat at constant pressure c Dg Binary diffusion coefficient Activation energy E Heat of gasification at the jump tem-I. perature Le Lewis number Ratio of specific heats cg/c r T Temperature Time t Surface regression rate Space coordinate in the condensed × phase v Mass fraction Y Reduced mass fraction defined in Eq. (24) Dimensionless inverse heat of gasifia cation c(Tgo-T1)/L Dimensionless activation energy de-8 fined in Eq. (26) Ratio of thermal responsivities Г  $\sqrt{(\rho \lambda c)_g/(\rho \lambda c)}$ Dimensionless parameter  $\alpha(r+\Gamma)$ ٨ Similarity variable 5/2/1 Dimensionless temperature θ  $(T-T_1)/(T_{go}-T_1)$ Thermal conductivity E, Dimensionless space coordinate defined in Eq.(12) Density Reduced time  $\beta^2 \tau / \Gamma^2$
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# Subscripts

- F Fuel
- g Gas
- 0 Oxidizer
- s Surface
- o Initial value
- Jump value.

#### I. Introduction

During the past years a considerable effort has been devoted towards understand ing the ignition mechanism of a condensed material. A large amount of data has been gathered experimentally and several theo-ries have been proposed to interpret these data. Unfortunately, however, ignition is a very complex phenomenon and many different physical and chemical processes are involved during an ignition experiment. The analytical tools available are not suited to tackle the complete problem and several simplifications have to be introduced. addition, some of the physical and chemical parameters involved in the ignition event are not known in a given experimental setup and this prevents meaningful comparisons with experimental data. Actual ly, these data are often used to derive the unknown parameters, thereby obtaining good agreement with those particular data.

At the present time, no consensus exists as to the location of the reaction responsible for ignition. In fact, one of the main objectives of both theoreticians and experimentalists working on ignition research has been to elucidate where for a class of propellants ignition takes place. To achieve this goal it is necessary to develop analysis with the same assumptions but differing only on the location of the key reaction leading to ignition. It appears that approximate analyses yield a better understanding of the phenomenon under study than a large amount of numerical results, although the accuracy of the latter for a given set of parameters is superior to that of the approximate methods. However, asymptotic techniques have been developed to a point such that while possessing good accuracy they still can retain as general assumptions as those used in the usual numerical analyses. These assumptions should be as real istic as possible and yet simple enough to be treatable by asymptotic techniques, and furthermore they should be amenable to be experimentally reproduced.

The results of these asymptotic analyses should be checked with good numerical solutions and then an experiment, reproducing as closely as possible the assumptions of the analyses should be carried out to obtain not only the ignition time but as many significant variables (such as temperature and mass fractions profiles) as possible. Only then meaningful experimental comparisons will be possible in trying to ascertain the location of the reaction producing ignition.

The ignition event starts with the application of a stimulus that enhances the gasification of the material and a selfacceleratory exothermic chemical process. Asymptotic analyses have been carried out assuming the same ignition stimulus, namely a constant flux of radiant energy absorbed it the surface but considering that either a condensed phase 1, heterogeneous 2 or gas-phase 3,4,5 exothermic reaction is responsible for ignition. We are developing similar analyses for an ignition stimulus consisting in the instantaneous contact of the condensed material with a hot This ignition stimulus can be reproduced experimentally by reflecting a shock wave on a condensed material located at the end of a shock tube. Interest in studying this particular type of ignition stems from the considerable amount of existing experiments 6-12, numerical results 13-17 and approximate analyses 6,11,18,19 that will facilitate the task of deriving conclusions regarding the ignition mechanism in a particular material. We have carried out recently asymptotic analyses for large activation energies of the heterogeneous 20 and condensed phase 21 shock-tube ignition, with the idea of generating simple analytical descriptions of the ignition process, and the present paper is a first step in a parallel study of gas-phase ignition. As pointed out previously<sup>3</sup>, gas-phase ignition theory is more complicated than either the heterogeneous or condensed phase theories, since it is necessary to consider two different chemical processes; gasification and ignition. The ignition event will be controlled by either of these sequential proc esses depending on their relative rates. The analysis of the gasification process reveals several interesting features which makes it advantageous to present separately such an analysis, to be followed by the study of the gas phase ignition process.

The gasification reaction may be considered as a rate process or as an equilibrium process. Usually equilibrium will be maintained only in the case of liquids. The analysis in the main text assumes a rate process described by a zero order endother mic Arrhenius rate law. However in an appendix we develop a parallel analysis for the equilibrium case, when the Clasius-Clapeyron equation is used to relate the surface mass fraction of gasified liquid to the surface temperature.

In the analysis we take into account

the convective effects due to the flow resulting from the gasification of the condensed material, together with the unsteady effects of heat conduction in the solid and gas phases and diffusion in the gas phase. The conservation equations are solved by asymptotic techniques based on the realistic assumption that the nondimensional activation-energy of the gasification reaction, or ratio of activation temperature to the solid initial temperature, is large compared to unity. In the context of this asymptotic analysis an integral equation is derived whose solution yields the surface temperature as a function of time; the sur face temperature rises instantaneously at t = 0 to a jump value, and then decreases slowly as a result of the endothermic gasi fication process which in addition blows the hot gases away from the solid surface thus reducing the heat conduction flux. Asymptotic expressions giving the temperature and mass fraction profiles for small and large values of time are also given in the paper.

## II. Formulation

We consider the case in which a condensed material at a temperature  $T_{\rm O}$ , occupies the half space x < 0. At time zero the temperature of the adjacent stagnant gas that occupies the half space x > 0 is suddenly raised to a high value  $T_{\rm go}$  to produce a corresponding rise in surface temperature, and thereby enhance a gasification reaction at the surface. Figure 1 shows the process so unted for in this study. As stated introduction, only gasification we can analyzed here and therefore the gasecular phase will be considered

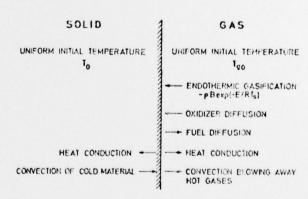


Fig.1 Schematic representation of the gasification process.

as inert. For convenience, the origin x=0 is maintained at the surface of the regressing material and the mass coordinate  $\psi$  is introduced in analyzing the gas phase

We will use the well justified assumptions that the pressure is constant and that the effect of external forces and viscous stresses are negligible. We use Fick's law to calculate the diffusion ve-

locities and consider the specific heats to be constant in each phase. It is also assumed that the density and thermal conductivity in the condensed phase as well as their product in the gas phase are constant. Under these conditions the conservation equations for energy in the solid and gas phases, and species conservation equations for oxidizer and fuel, are respectively

$$\rho c \frac{\partial T}{\partial t} + \rho v c \frac{\partial T}{\partial x} = \frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right)$$
 (1)

$$2g \frac{\partial T}{\partial t} + \rho v 2g \frac{\partial T}{\partial \psi} = \frac{\partial}{\partial \psi} \left( \rho_g \lambda_g \frac{\partial T}{\partial \psi} \right) \tag{2}$$

$$\frac{\partial Y_F}{\partial t} + \rho v \frac{\partial Y_F}{\partial \psi} = \frac{\partial}{\partial \psi} \left( \rho_g^2 D_g \frac{\partial Y_F}{\partial \psi} \right)$$
 (3)

An equation identical to (3) exists for the oxidizer mass fraction  $Y_0$ , except that the oxidizer diffusivity  $D_{go}$  should replace  $D_g$ . The initial and boundary conditions are

$$T(0,x)=T(t,-\infty)=T_{o}$$
,  $T_{g}(0,\psi)=T_{g}(t,\infty)=T_{go}$ , 
$$T(t,0)=T_{g}(t,0)$$
 (4

$$Y_{F}(0,\psi) = Y_{F}(t,\infty) = 0, Y_{O}(0,\psi) = Y_{O}(t,\infty) = Y_{O\infty}$$
 (5)

$$-\lambda \left. \frac{\partial T}{\partial x} \right|_{S} + \rho_{g} \lambda_{g} \left. \frac{\partial T}{\partial \psi} \right|_{S} = \rho v \left[ \left( c_{g} - c \right) \left( T_{S} - T_{1} \right) + L \right]$$
 (6)

$$\rho v Y_{OS} = \rho_g^2 D_{go} \frac{\partial Y_O}{\partial \psi} \bigg|_{S}$$
 (7)

$$\rho v \left( Y_{F_S} - 1 \right) = \rho_E^2 D_g \left. \frac{\partial Y_F}{\partial \psi} \right|_S \tag{8}$$

where all the symbols are defined in the nomenclature. The surface regression rate  ${\bf v}$  is given by

$$v = Bexp(-E/RT_s)$$
 (9)

and L is the heat of gasification referred to the "jump" temperature  $T_1$ , which is the one established at the surface immediately after the application of the hot gases. In absence of gasification the surface temperature remains fixed at its "jump" value

$$T_1 = T_0 + \left(T_{go} - T_0\right) \frac{\Gamma}{1 + \Gamma} \tag{10}$$

In the analysis that follows, the quantity  $\rho_g \lambda_g$  is taken as constant, and its value is chosen in such a way that the correct "jump" temperature (with arbitrary dependence of  $\rho_g \lambda_g$  on temperature) calculated in reference 20 is recovered, when the inert problem is solved in the assumption of constant thermal properties.

Introducing 'the nondimensional variables

$$\theta = \frac{T-T_1}{T_{go}-T_1}$$
 ,  $\theta_g = \frac{T_g-T_1}{T_{go}-T_1}$  (11)

$$\xi = \begin{cases} \frac{L}{T_{go}^{-}T_{1}} \frac{\rho v_{1}}{\rho_{g}\lambda_{g}} \frac{\Gamma}{1+\Gamma} \psi & \text{in the gas phase} \\ \\ -\frac{\rho v_{1}L}{T_{go}^{-}T_{1}} \sqrt{\frac{\rho c}{\rho_{g}\lambda_{g}^{-}c_{g}\lambda}} \frac{\Gamma}{1+\Gamma} \times & \text{in the solid phase} \end{cases}$$

$$\tau = \left(\frac{L}{T_{go}-T_1}\right)^2 \frac{(\rho v_1)^2}{\rho_g \lambda_g c_g} \left(\frac{\Gamma}{1+\Gamma}\right)^2 t \qquad (13)$$

where  $v_1 = Bexp(-E/RT_1)$  is the value of v for  $T_S = T_1$ , equations (1)-(8) become

$$\frac{\partial \theta}{\partial \tau} - \alpha \left(1 + \Gamma\right) \frac{\mathbf{v}}{\mathbf{v_1}} \frac{\partial \theta}{\partial \xi} = \frac{\partial^2 \theta}{\partial \xi^2} \tag{14}$$

$$\frac{\partial \theta_{g}}{\partial \tau} + r\alpha \frac{1+\Gamma}{\Gamma} \frac{\mathbf{v}}{\mathbf{v}_{1}} \frac{\partial \theta_{g}}{\partial \xi} = \frac{\partial^{2} \theta_{g}}{\partial \xi^{2}}$$
 (15)

$$\frac{\partial Y_F}{\partial \tau} + r\alpha \frac{1+\Gamma}{\Gamma} \frac{v}{v_1} \frac{\partial Y_F}{\partial \xi} = \frac{1}{Le} \frac{\partial^2 Y_F}{\partial \xi^2}$$
 (16)

$$\theta(0,\xi) = \theta(\tau,\infty) = -\Gamma$$
,  $\theta_g(0,\xi) = \theta_g(\tau,\infty) = 1$ ,

$$\theta(\tau,0) = \theta_{g}(\tau,0) \tag{17}$$

$$Y_{F}(0,\xi) = Y_{F}(\tau,\omega) = 0$$
,  $Y_{0}(0,\xi) = Y_{0}(\tau,\omega) = Y_{0\omega}$  (18)

$$\frac{\partial \theta}{\partial \xi} \bigg|_{S} + \Gamma \frac{\partial \theta}{\partial \xi} \bigg|_{S} = (1+\Gamma) \frac{L + (T_{S} - T_{1}) (c_{g} - c)}{L} \frac{v}{v_{1}}$$
(19)

$$Y_{0s} \operatorname{Lear} \frac{1+\Gamma}{\Gamma} \frac{v}{v_1} = \frac{\partial Y_0}{\partial \xi} \bigg|_{S}$$
 (20)

$$(Y_{Fs}-1)$$
 Lear  $\frac{1+\Gamma}{\Gamma} \frac{v}{v_1} = \frac{\partial Y_{Fs}}{\partial \xi} \bigg|_{S}$  (21)

where  $v/v_1 = exp(-E/RT_s+E/RT_1)$ .

An equation identical to (16) defines the oxidizer mass-fraction  $Y_0$ , provided that the Lewis number for the oxidizer Le  $_0$  is used instead of Le.

# III. Surface Temperature and Regression Rate History.

At small times the temperature gradients at the surface are large and therefore the energy absorbed by the gasification reaction is negligible compared to the energy transport by conduction from the gas phase to the condensed phase. Upon contact with the hot gases the surface tem perature will be instantaneously raised to a jump value corresponding to zero gasification rate but, subsequently, gasification introduces two effects that cause the surface temperature and the regression rate to decrease with time. First, a fraction of the energy coming from the gas phase is absorbed by the gasification reaction, so that the surface temperature can not be maintained at its jump value. Second, the convective flow resulting from the condensed phase gasification blows the hot gas away from the surface and brings cold material towards the surface, thereby reducing the surface temperature and the regression rate.

We shall analyze first the period of time when the surface regression rate is reduced by a factor of order unity from its "initial" value corresponding to the jump temperature  $T_1$ . For large values of the non-dimensional activation energy E/RT1, small increments in the surface temperature of the order RT $_1^2/\mathrm{E}$  produce a change in the gasification rate by a factor e. For those early times the corresponding deviations of  $\theta$  and  $\theta_g$  from their inert values

$$\theta_{Ig} = -\theta_{I}/\Gamma = erf(\xi/2\sqrt{\tau}) \tag{22}$$

are small, of order  $RT_1^2/\{E\{T_{go}-T_1\}\}$ , so that it is convenient to describe this early period in terms of the non-dimensional variables

$$\chi = \beta(\theta_1 - \theta)/\Gamma$$
 ,  $\chi_g = \beta(\theta_{1g} - \theta_g)/\Gamma$  (23)

$$y = \beta Y_{F} / \left[ \alpha r (1+F) \sqrt{Le} \right]$$
 (24)

$$\sigma = \beta^2 \tau / \Gamma^2$$
 ,  $z = \beta \xi / \Gamma$  (25)

where

$$\beta = E\left(T_1 - T_0\right) / RT_1^2 \tag{26}$$

is a non-dimensional activation energy. The new non-dimensional time  $\sigma$  and fuel mass fraction y have been defined so that they are of order unity during this period. The analysis that follows is based on the assumption that the parameter  $\beta$  is large compared to unity which is equivalent to require that the gasification rate at the jump temperature be much faster than the gasification rate at the initial temperature. Thus, it is necessary to assume that B is large in order to have a physically realistic problem in which a non reacting condensed material is suddenly brought to vigorous gasification upon contact with hot gases. The ratio of thermal responsivities  $\Gamma$  is usually small, but it may become of order unity for large values of pressure or T , since it is proportional to the square root of  $\rho_{g}\lambda_{g}$  . Thus, the parameter  $\beta/\Gamma$  is at least of order  $\beta$ .

Introducing the non-dimensional variables (23)-(25) and taking the limit  $\beta \to \infty$ , equations (14)-(21) simplify to:

$$\frac{\partial \chi}{\partial \sigma} - \frac{\partial^2 \chi}{\partial z^2} = \frac{P}{\sqrt{\pi \sigma}} \exp \left(-\frac{z^2}{4\sigma} - \chi_s\right) \tag{27}$$

$$\frac{\partial y}{\partial \sigma} - \frac{1}{Le} \frac{\partial^2 y}{\partial z^2} = 0 \tag{28}$$

$$\chi(0,z) = \chi(\sigma,\infty) = y(0,z) = y(\sigma,\infty) = 0$$
,

$$\chi(\sigma,0) = \chi_{g}(\sigma,0) \tag{29}$$

$$\frac{\partial \chi}{\partial z}\Big|_{S} + \Gamma \frac{\partial \chi_{g}}{\partial z}\Big|_{S} = -(1+\Gamma)\exp(-\chi_{S})$$
 (30)

$$\frac{\partial y}{\partial z}\Big|_{S} = -\sqrt{\text{Le}} \exp(-\chi_{S})$$
 (31)

The factor exp  $(-\chi_S)$  =  $v/v_1$  accounts for the reduction of gasification rate with temperature.

The parameter P appearing in equation (27) for the case of condensed phase temperature  $\chi$  equals  $\alpha\Gamma(1+\Gamma)$ . The equation for the gas phase temperature  $\chi_g$  is identical to Eq.(27) with P replaced by  $\alpha\Gamma(1+\Gamma)/\Gamma$ .

Only the gradient of the inert temperature appears in the effect of convection given by the right-hand side of Eq (27), and no convective effects appear in Eq (28) describing the mass fractions. It should also be observed that in the limit  $\beta \to \infty$  the equations and boundary conditions describing the temperature distribution are uncoupled from those describing mass fraction. Green functions may be used to write the solution to equations (27), (29) and (30) as

$$\chi(\sigma,z) = \frac{P}{2\pi} \int_{0}^{\sigma} \frac{\exp(-\chi_{s}^{\prime})}{\sqrt{\sigma'(\sigma-\sigma')}} \int_{0}^{\infty} \exp(-z^{\prime 2}/4\sigma') \times$$

$$+ \int_{0}^{\sigma} \frac{\exp\left[-z^{2}/4(\sigma-\sigma')\right]}{\sqrt{\pi(\sigma-\sigma')}} \left(-\frac{\partial \chi'}{\partial z}\Big|_{S}\right) d\sigma' \qquad (32)$$

where

$$G(z,\sigma; z',\sigma') = \exp\left[-(z-z')^2/4(\sigma-\sigma')\right] + \exp\left[-(z+z')^2/4(\sigma-\sigma')\right]$$
 (33)

Evaluating equation (32) at the surface and using equation (30) an integral

equation defining x is obtained, viz.,

$$\chi_{s} = \frac{\Delta}{\sqrt{\pi\sigma}} \int_{0}^{\sigma} \exp(-\chi_{s}^{*}) d\sigma^{*} + \int_{0}^{\sigma} \frac{\exp(-\chi_{s}^{*})}{\sqrt{\pi(\sigma - \sigma^{*})}} d\sigma^{*}$$
(34)

This equation describes the evolution with time of the surface temperature as a function of the unique parameter  $\Delta=\alpha(r+\Gamma)$  , which is a measure of the cooling effect of the gasification flow. The second term in the right hand side of equation (34) accounts for the heat absorbed by the endothermic gasification and will be the only one appearing if the convective effects were neglected.

From equations (28), (29) and (31) an integral equation may be derived relating the surface mass fraction to time, viz.,

$$y_s = \int_0^{\sigma} \frac{\exp(-\chi_s')}{\sqrt{\pi(\sigma - \sigma')}} d\sigma'$$
 (35)

This equation can also be used to calculate the surface value of the oxidizer mass fraction, if we use  $1-Y_0/Y_{0\infty}$  instead of  $Y_\Gamma$  in the definition of y (Eq.24). It is observed that the mass fractions at the surface are proportional to the second term in the right hand side of (34) associated with the gasification reaction, since in the limit  $\beta \to \infty$ , the convective effects are negligible.

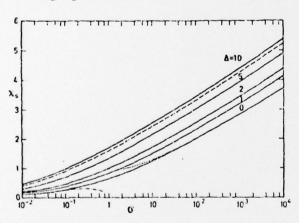


Fig. 2 Surface temperature  $\chi_s = (T_1 - T_s) E/RT_1^2$  as a function of time  $\sigma$ , and  $\Delta$ .

A finite difference version of equations (34) and (35) has been numerically integrated through a procedure paralleling that used in reference 1. The results of such an integration are shown in figures (2) and (3) for several values of  $\Lambda$ . The non-dimensional regression rate  $v/v_1$  =  $\exp(-\chi_S)$  is plotted in figure (4). This rate is maximum at the time of contact with the hot gas and continuously decreases towards zero afterwards.

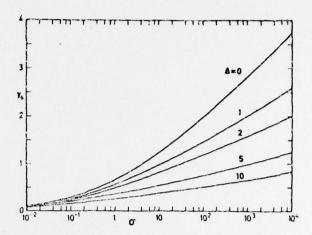


Fig. 3 The function y  $(\sigma, \Lambda)$  describing surface mass fraction history.

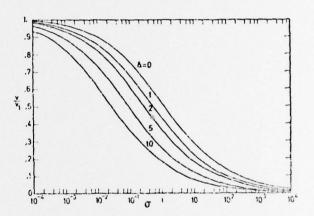


Fig. 4 Surface regression rate history.

For large values of  $\Delta$ , the characteristic non-dimensional time is  $\sigma_1=\Delta^2\sigma$ . Introducing this new variable in equation (34) and taking the limit  $\Delta+\infty$ , the last term of Eq.(34) disappears and the following differential equation is derived

$$\frac{\mathrm{d}\chi_s}{\mathrm{d}\sigma_1} = \frac{\exp(-\chi_s)}{\sqrt{\pi\sigma_1}} - \frac{\chi_s}{2\sigma_1}$$
 (36)

A Runge-Kutta procedure was used to solve euqation (36). The dashed curve in figure (2) represents  $\chi_{\rm S}$  as a function of a for  $\Delta$  = 10 obtained from equation (36), showing as expected, differences of order  $1/\Delta$  with the exact solution. This curve and the definition of  $\sigma_1$  may be used to obtain  $\chi_{\rm S}(\sigma_1)$ . In this limit, the convective effec appearing in equation (27) has to be retained, but the right hand side of equation (30) associated with the heat absorbed by gasification, is negligible.

Since one of the potentital uses of the

present study stems from its application to gas phase ignition, it is interesting to observe that under certain particular conditions, the present analysis can be easily generalized to account for fast gas phase reactions and even to describe the ignition event. Namely, if the fuel reacts immediately after gasification with the ambient oxidizer, the heat released by this exothermic reaction should be subtracted from the heat absorbed by the gasification reaction L. It is not difficult to show that in this case  $Y_F = 0$  in the gas phase and the ratio  $(Y_{0\infty}-\dot{Y}_0)/(Y_{0\infty}+\nu)$ , where  $\nu$  is the mass stoichiometric ratio oxidizer/fuel, satisfies Eq (3) and the same initial and boundary condition than the fuel mass frac tion in the non-reactive case. In particular if the net value of L is positive Fig. 3 giving  $y_s(\sigma)$ , can be used to calculate  $Y_{OS}(\sigma)$  if we write  $(Y_{OS}-Y_{OS})/(Y_{OS}+\nu)=y_s\alpha r(1+\Gamma)\sqrt{Le}/\beta$ . The analysis ceases to be valid if the resulting value Yos becomes negative, when the reaction zone moves away from the surface.

For sufficiently exothermic reactions the net value of L is negative; under these conditions,  $\Delta$  is negative and the surface temperature satisfies Eq .(34) if its left hand side is replaced by  $(-\chi_S)$ . This equation has been numerically integrated in reference 20,tc generate  $\sigma_{ign}$  as a function of  $\Delta$ . It is observed that ignition occurs only for values of  $-\Delta < 2$ , in which case  $-\chi_S$  is positive and grows to infinite at a finite time  $\sigma_{ign}(\Delta)$ .

# IV. Temperature and Concentration Profiles

The temperature and concentration profiles can be calculated, in principle, by using Eq.(32) in terms of the calculated surface temperature history. However, we shall instead, derive expansions for the temperature and concentration profiles for small and large values of the non-dimensional time  $\sigma$ . In these expansions the non-dimensional distance z to the solid surface is scaled with the characteristic thickness of the heat conduction layer  $2\sqrt{\sigma}$  to produce the similarity variable  $\eta=z/2\sqrt{\sigma}$  for small and large times. We shall later use these expansions to generate approximate expressions that can be used to calculate the surface temperature for all times.

For small values of  $\sigma$  the factor  $\exp(-\chi_s)$  in Eq.(27), accounting for the change in regression velocity with changes in surface temperature, can be written equal to 1 and, in addition, the heat of gasification in Eq.(30) can be neglected; the resulting system of equations has a similarity solution of the form  $\chi = \sqrt{\sigma f_o}(\eta)$ , where

$$f_0 = [(2+\Delta)/\sqrt{\pi}] \exp(-\eta^2) + (P-2-\Delta) \operatorname{nerfcn}$$
 (37)

In order to obtain a more accurate expression of  $\chi$  for small  $\sigma$  we can use the expansion

$$\chi = \sqrt{\sigma} f_0 + \sigma f_1 + --- \tag{38}$$

which, when substituted in Eqs.(27), (29) and (30), previously written in terms of the variables  $\sigma$  and  $\eta$  leads to a system of equations, with solutions given by Eqs.(37) and

$$f_1 = \left[ -2 - \Delta + \frac{4(\Delta + 2)(P - \Delta)}{3\pi} \right] \left[ \frac{\text{erfcn}}{2} + \eta^2 \text{erfcn} \right]$$
$$- \frac{\text{nexp}(-\eta^2)}{\sqrt{\pi}} - \frac{2P}{3\pi} (\Delta + 2) \exp(-\eta^2)$$
(39)

where P equals  $\alpha\Gamma(1+\Gamma)$  in the condensed phase and  $\alpha\Gamma(1+\Gamma)/\Gamma$  in the gas phase.

Thus, for early times the surface temperature is given by

$$\chi_{s} = \sqrt{\frac{\sigma}{\pi}} \left(2+\Delta\right) - \sigma \left[1 + \frac{\Delta}{2} + \frac{2\Delta}{3\pi} \left(\Delta + 2\right)\right] + ---(40)$$

This expansion can also be derived directly from equation (34). It is plotted for comparison with the exact solution in figure (2) for the case  $\Delta=0$ . Similarly, the early times mass fraction profiles are given by

$$y = 2\sqrt{\sigma} \left[ \frac{\exp(-\mu^2)}{\sqrt{\pi}} - \operatorname{perfc}(\mu) \right]$$

$$-\sigma(2+\Delta) \left[ \mu^2 \operatorname{errc}(\mu) - \frac{\mu}{\sqrt{\pi}} \exp(-\mu^2) + \frac{\operatorname{erfc}(\mu)}{2} \right] + --- \tag{41}$$

where  $\mu = \eta \sqrt{Le}$ .

For large  $\sigma$  the surface value  $\chi_s$  of  $\chi$  is large and it becomes a slowly varying function of  $\sigma$ . If Eqs. (27) and (28) are written in terms of the similarity variable  $\eta$  and  $\sigma$ , the time derivative term in the equations can be neglected in first approximation for large times, and the temperature and concentration profiles take a quasi-similarity form. A more accurate representation of the temperature and concentration profiles can be obtained as expansions of  $\chi$ ,  $\chi_g$  and  $\gamma$  in decreasing powers of  $\chi_s$  times functions of the variable  $\eta$ . Thus when the expansion

$$\chi = \chi_s(\sigma)F_o(\eta) + F_1(\eta) + (1/\chi_s)F_2(\eta) + ---$$

and a similar expansion in powers of  $\chi_g$  for  $\chi_g$  are introduced in Eq.(30), the

relation

$$\frac{\sqrt{\sigma}}{\chi_{s} \exp(\chi_{s})} = a_{o} + \frac{a_{1}}{\chi_{s}} + \frac{a_{2}}{\chi_{s}^{2}} + --- \tag{43}$$

is obtained, where

$$a_{i} = -\frac{F'_{i}(0) + FF'_{ig}(0)}{2(1+\Gamma)}$$
 (44)

and the subscript g indicates gas phase variable. When Eqs(27) and (29) are also written in terms of  $\eta$  and  $\chi_s$  and  $\chi$  and  $\chi_s$  are substituted by their expansion (42) we obtain, by collecting like powers of  $\chi_s$ , a system of equations with the solutions

$$F_o = \operatorname{erfc}(\eta) + \frac{2P}{\sqrt{\pi}} a_o \left[ \exp(-\eta^2) - \operatorname{erfc}(\eta) \right]$$
 (45)

$$F_{1} = \left[\overline{g}(\eta) - \overline{g}(\infty) \operatorname{erf} \eta\right] \left[1 - \frac{2 \operatorname{Pa}_{0}}{\sqrt{\pi}}\right] + \frac{2 \operatorname{P}}{\sqrt{\pi}} \left(a_{1} - a_{0}\right) \left[\exp(-\eta^{2}) - \operatorname{erfc}(\eta)\right]$$
(46)

where the function  $\overline{g}(x)$  is the solution of the equation

$$\overline{g}'' + 2x\overline{g}' = 2 \operatorname{erfc}(x)$$
,  $\overline{g}(0) = 0$ ,  $\overline{g}'(0) = 1$  (47)

shown with a dashed line in figure (5). We notice for future reference that  $\overline{g}(\infty) \approx 1.58$ .

The expression for  $-F_2$  is identical to that of  $F_1$  with  $a_1$  replaced by  $-a_2$ . Equation (44) can now be used to calculate the unknown parameters  $a_1$ , thereby obtaining

$$a_0 = \sqrt{\pi/(\pi + 2\Delta)} \tag{48}$$

$$a_1 = -a_2 = \left(\frac{2\Lambda}{\pi^{3/2}} - \frac{1}{2} + \frac{\overline{g}(\infty)}{\sqrt{\pi}}\right) \left(1 + \frac{2\Lambda}{\pi}\right)^{-2}$$
 (49)

The asymptotic expansion in Eq.(43) can be used to calculate  $\chi_s(\sigma)$  for large times. The relation obtained when using the first three terms of the expansion has been plotted in figure 2, for  $\Delta=0$ , to show the range of values of  $\sigma$  when it is an accurate representation of the exact solution.

A similar procedure yields the long time mass fraction profiles, written here in terms of the similarity variable  $\mu=\eta\sqrt{Le}$ 

$$y = \lambda_s a_o \sqrt{\pi} \operatorname{erfc}(\mu) + \left\{ a_o \sqrt{\pi} \left[ \overline{g}(\mu) - \overline{g}(\infty) \right] + \sqrt{\pi} \left( a_1 + \frac{\sqrt{\pi} a_o}{2} \right) \operatorname{erfc}(\mu) \right\} \left( 1 - \frac{1}{\chi_s} \right) + --- \quad (50)$$

It is observed that the mass fraction profiles depend only on the parameter  $\boldsymbol{\Delta}$ .

Equations (40) and (43) may be used to derive an approximate form of the relation  $\sigma(\chi_{\kappa})$  valid for all times.

$$\sigma = a_0^2 \chi_S^2 \exp(2\chi_S) \frac{\chi_S^2 + a\chi_S - b + d_1 + d_2\chi_S}{\chi_S^2 + d_3 + d_4\chi_S}$$
 (51)

where

$$a = \frac{2a_1}{a_0}$$
 ,  $b = \frac{2a_1}{a_0} - \frac{a_1^2}{a_0^2}$  (52)

$$d_{2} = d_{1} = \frac{bf - 2gb + ag - aa_{0}^{2}}{a\left[f - 2g - \frac{2g}{a} + \frac{a_{0}^{2}}{a} + \frac{g^{2}}{aa_{0}^{2}}\right]}$$
(53)

$$d_3 = d_1 - d_4 a = \frac{aa_0^2 + ba_0^2/a - gb/a}{a\left[f - 2g - \frac{2g}{a} + \frac{a_0^2}{a} + \frac{g^2}{aa_0^2}\right]} (54)$$

$$f = \frac{\pi^2(1+4\Delta/3\pi)}{(2+\Delta)^3}$$
,  $g = \pi/(2+\Delta)^2$  (55)

Expression (51) when expanded for large  $\sigma$ , reproduces the first three terms of equation (43), and when expanded for small  $\sigma$ , reproduces the correct expansion up to terms of order  $\chi_s^3$ . The approximate relation (51) when drawn in figure (2) can not be distinguished from the exact solution obtained numerically.

Since the objective of the present study is the analysis of the gasification process in order to develop a follow up investigation of gas phase ignition, we are specially interested in obtaining temperature and mass fraction profiles in the gas phase.

Equations (38) and (41) valid for small values of time and equations (42) and (50) valid for long times will permit the analysis of early ignitions (very reactive gas phase) and late ignitions, thereby covering both limiting behaviours.

In figure (5) profiles of temperature deviations  $\chi$ ,  $\chi_g$  from the inert or nongasification values and the non-dimensional mass fraction  $\chi$  are shown as a function of  $\eta$  for different values of  $\sigma$ . We have only shown the case  $\alpha=0$ , Le = 1, for which the equations and boundary conditions defining  $\chi$ ,  $\chi_g$ , and  $\chi_g$  coincide. Using figure (5) and equations (22), (23), it is easy to derive the temperature profiles for a given  $\beta/\Gamma$ .

Notice that the fuel mass fraction does not become of order unity until very long times when  $\chi$  becomes of order  $\beta$  , so

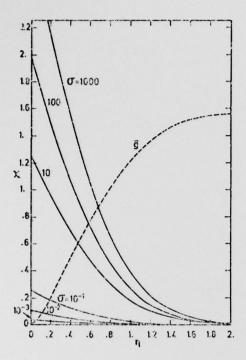


Fig. 5 Temperature and mass fraction profiles for  $\alpha = 0$ , Le = 1.

so that

$$= (T_1 - T_s) (T_1 - T_o)$$
 (56)

is of oracle antry, and the solution given by Eqs.(42) and (50) is no longer valid. To obtain the solution of the system of Eqs.(14)-(21) for those large values of time, we write the system in terms of the variables  $\eta = \xi/2\sqrt{\tau}$  and  $\tau$ , and notice that the time derivative terms are of order 1/8 relative to the remaining terms. Thus, in first approximation we can use the quasi-similarity assumption as we did when founding the solution of the system of Eqs.(27)-(30) for large  $\sigma$ .

The resulting system of ordinary differential equations can be solved to yield

$$(\theta_T - \theta)/\Gamma = Aerfc(\eta + u\Gamma/r) + erfc\eta$$
 (57)

$$(\theta_{Ig} - \theta_g) = Berfc(\eta - u) - erfc\eta$$
 (58)

where

$$u\Gamma/\{r\alpha(1+\Gamma)\sqrt{\tau}\} = v/v_1. \qquad (59)$$

$$v/v_1 = exp\{-s\phi_s/[1+\phi_s(1-T_0/T_1)]\}$$
 (60)

and the constant A and B are given by the equations

$$\phi_{s} = Aerfc(\Gamma u/r)+1 = \{Perfc(-u)-1\}/\Gamma$$
 (61)

$$A \exp \left\{ -(Tu/r)^{2} \right\} + B \exp \left( -u^{2} \right) =$$

$$= \sqrt{\pi} u \left\{ 1 - \Phi_{s} \left( T_{1} - T_{0} \right) \left( c_{g} - c \right) / L \right\} / \alpha r \qquad (62)$$

obtained from the boundary conditions in Eqs.(17) and (19).

Eqs.(61) and (62) give A, B and  $\Phi_g$  explicitely in terms of u, while Eq.(59) gives  $\tau(u)$ . Thus, we have an explicit parametric representation of the relation  $\Phi_g(\tau)$  involving the parameter u. Since u is of order unity when  $\Phi_g$  is of order unity, an explicit relation giving  $\Phi_g(\tau)$ , with errors of order 1/3, cen be obtained by writing u = 1 in Eq.(59).

The fuel mass fraction is given by

$$Y_{F}/Y_{Fs} = \frac{\text{erfc}\{(\eta - u)\sqrt{\text{Le}}\}}{1 + \text{erf}(u\sqrt{\text{Le}})}$$
(63)

where Y<sub>Fs</sub> is determined by

$$Y_{FS} = 1 - \frac{Y_{FS}}{u\sqrt{\pi Le}} \frac{\exp(-u^2 Le)}{1 + \exp(u\sqrt{Le})}$$
 (64)

obtained from the boundary condition in Eq.(21). A similar equation is obtained to describe the exidizer mass fraction by replacing  $Y_F$  and  $Y_{FS}$  by 1- $Y_0/Y_{0\infty}$ , and 1- $Y_{0S}/Y_{0\infty}$  respectively.

## V. Conclusions

The time evolution of the temperature and mass fractions at the surface of a condensed material that gasifies according to an endothermic Arrhenius decomposition reaction after being exposed to a hot gas, has been described analytically by means of asymptotic techniques.

Upon contact with the hot gas, the sur face temperature is raised to an intermediate "jump" value equal to the one existing in absence of gasification. Gasification however, causes the surface temperature to decrease with time, since part of the energy coming from the hot gas is absorbed by the endothermic reaction and, in addition, because the gasification flow brings cold material towards the surface and blows the hot gases away. During an early period in which the decreasing surface regression rate is still of the order of its initial value at the jump temperature, the Arrhenius exponent can be linearized around the jump temperature  $T_1$  and only the gradient of the non-gasification temperature profile enters the convective effects. As a result of these simplifications, the surface temperature history can be described by the solution of an integral equation giving  $\chi_s = E(T_1 - T_s)RT_1^2$  in

terms of a non-dimensional time  $\sigma$  and a single parameter  $\Delta$  measuring the convective effects; this is represented in Fig.2. The surface value of the fluid mass fraction is given in Fig.3. Closed form expressions of the temperature and concentration profiles are also given in the form of expansions for small and large values of  $\sigma$ .

When the variable  $\chi_s$  becomes large of order  $\beta$ , the complete convective terms have to be retained and the exact form of the Arrhenius exponent has to be used. However, for those large times a quasissimilar solution exist when the distance to the surface is scaled by  $2\sqrt{\tau}$ , so that explicit expressions have been obtained for the temperature and concentration profiles in terms of the similarity variable  $\eta$  and a function u of time given by Eq. (59).

We consider in an Appendix the case when the gasification is assumed to obey an equilibrium Clasius-Clapeyron condition instead of the regression rate law of Eq. (9). The temperature and mass fraction profiles are described in terms of a similarity solution valid for all times, involving the similarity variable  $\eta=\xi/2\sqrt{\tau}.$  The surface temperature does not change with time after jumping at  $\tau=0$  to a value determined by gasification effects. The regression rate is found to vary as  $1/\sqrt{\tau}$ .

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## Appendix

In this appendix we present an analysis of the cases when phase equilibrium is maintained during the gasification process. In practice the equilibrium analysis will be usually applicable to the case of liquid fuels and the rate process analysis will be better suited for the case of solid propellants, plastics and other solid materials.

The Clasius-Clapeyron equation relates the surface mass fraction to the surface temperature through

$$Y_{FS} = \exp - \int_{T_S}^{T_B} \frac{LdT}{RT^2}$$
 (A1)

where we have made the non-essential assumption that the molecular weights of the gaseous species are equal. If in addition L changes slowly in the range of integration, then

$$Y_{FS} = \exp\left[L/RT_B - L/RT_S\right]$$
 (A2)

Equation (1)-(8) remain valid, but equation (6) is replaced by

$$-\lambda \left. \frac{\partial T}{\partial \chi} \right|_{S} + \rho_{g} \lambda_{g} \frac{\partial T_{g}}{\partial \psi} \right|_{S} = \frac{LD_{g} \rho_{g}^{2}}{Y_{FS} - 1} \frac{\partial Y_{F}}{\partial \psi} \bigg|_{S}$$
 (A3)

This problem is simpler than the one in the main text since it admits a similar ity solution valid for all times. It turns out that the regression velocity varies with time as  $1/\sqrt{\tau}$  and the temperature and concentration profiles become time independent if described in terms of the similarity variable  $\eta = 5/2\sqrt{\tau}$ . Introducing

$$V = \frac{\partial Y_F / \partial \eta}{2 \operatorname{Le}(Y_{FS}^{-1})} = \sqrt{\frac{c_g t}{\rho_g \lambda_g}} \rho v \tag{A4}$$

the solution to equations (1)-(8) is

$$\frac{T - T_o}{T_{go} - T_o} = C_1 erfc \left(n + \frac{\Gamma V}{r}\right)$$
 (A5)

$$\frac{T_g - T_o}{T_{go} - T_o} = 1 - C_2 \operatorname{erfc}(\eta - V)$$
 (A6)

$$Y_{\Gamma} = Y_{Fs} \frac{\text{erfc}[(n-V)\sqrt{Le}]}{\text{erfc}(-V\sqrt{Le})}$$
 (A7)

and the boundary conditions, yield

$$Y_{FS} = \left[1 + \frac{\exp(-V^2 \text{Le})}{V\sqrt{\pi \text{Le erfc}(-V\sqrt{\text{Le}})}}\right]^{-1}$$
 (A8)

$$\frac{T_s - T_o}{T_{go} - T_o} = C_1 \operatorname{erfc} \left(\frac{\Gamma V}{r}\right) = 1 - C_2 \operatorname{erfc} \left(-V\right) =$$

$$\frac{\text{re}^{-V^2}/\text{erfc}(-V) - V\Gamma\sqrt{\pi}/\text{ra'}}{\text{re}^{-V^2}/\text{erfc}(-V) + \exp\{-(\Gamma V/r)^2\}/\text{erfc}(\Gamma V/r)}$$
(A3)

where

$$\alpha' = \frac{c(T_{go} - T_o)}{L}$$
 (A10)

From equation (A9), the value of  $T_{\rm S}/T_{\rm O}$  is obtained as a function of V for fixed values of  $\alpha', r, \Gamma$ . Equation (A2) is then used to deduce  $T_{\rm B}/T_{\rm O}$  as a function of  $\alpha'$ , r,  $\Gamma$ , Le, L/RT $_{\rm O}$ , and  $T_{\rm gO}/T_{\rm O}$ . Once this curve is known, the value of V for the given  $T_{\rm B}$  is calculated, and therefore the value of  $T_{\rm S}/T_{\rm O}$  can be deduced from equation (A9). By this procedure the temperature and mass fraction distribution are known for all time.

Since the solution to the gasification problem involves several parameters, there by possessing many interesting limiting behaviours whose analysis will considerably lengthen the present paper, a parametric study of gasification under equilibrium conditions is postponed to a subsequent publication.

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profiles for small and large times. A closed form analytical description is also given for the gasification process under surface equilibrium \*Clasius Clapeyron\* conditions. In this case the surface temperature remains at a constant jump value, and the temperature and concentration prefiles have a similarity form.

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